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IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF :

HIROYUKI NAITOU, ET AL.

: EXAMINER: HAILEY, P. L.

SERIAL NO: 10/519,407 :

FILED: JANUARY 5, 2005

: GROUP ART UNIT: 1755

FOR: PROCESS FOR PRODUCING :
CATALYSTS FOR THE PRODUCTION
OF METHACRYLIC ACID

APPEAL BRIEF

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

SIR:

This is an appeal of the Final Rejection dated September 7, 2006 of Claims 1-15. A Notice of Appeal, along with a second-month extension of time, was timely filed on February 7, 2007.

I. REAL PARTY IN INTEREST

The real party in interest in this appeal is Mitsubishi Rayon Co., Ltd., having an address at 6-41, Konan-chome, Minato-ku, Tokyo 108-8506, Japan.

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II. RELATED APPEALS AND INTERFERENCES

Appellants, Appellants' legal representative and the assignee are aware of no appeals, interferences, or judicial proceedings which may be related to, directly affect or be directly affected by or have a bearing on the Board's decision in this appeal.

III. STATUS OF THE CLAIMS

Claims 1-15, all the claims in the application, stand rejected and are herein appealed.

IV. STATUS OF THE AMENDMENTS

No amendment under 37 CFR 1.116 has been filed, although a timely Request for Reconsideration after Final was filed. In an Advisory Action entered January 30, 2007, the Examiner refused to withdraw the rejection.

V. SUMMARY OF THE CLAIMED SUBJECT MATTER

Independent Claim 1 is drawn to a method of producing a catalyst having a particular formula $P_aMo_bV_cCu_dX_eY_fZ_gO_h$ (wherein X, Y, Z, and a-h are as defined), useful for subjecting methacrolein to vapor phase catalytic oxidization with molecular oxygen, the method comprising mixing

100 parts by mass of a solution or a slurry (liquid A), containing molybdenum atoms, phosphorous atoms and vanadium atoms, in which the content of ammonium species is 0 to 1.5 mol relative to 12 mol of the molybdenum atoms,

5 to 300 parts by mass of a solution or a slurry (liquid B), containing 6 to 17 mol of ammonium species relative to 12 mol of the molybdenum atoms contained in the solution A, and

a solution or a slurry (liquid C), containing an element Z,

wherein the liquid B is mixed with the liquid A, the liquid C or a mixture of the liquid A and the liquid C over a period of 0.1 to 15 minutes.

See original Claim 1, and the specification at page 2, line 4 to page 3, line 4.

VI. GROUNDS OF REJECTION

Ground (A)

Claims 1-15 stand rejected under 35 U.S.C. § 103(a) as unpatentable over JP 2000-296336A (Naito et al).

Ground (B)

Claims 1-15 stand rejected under 35 U.S.C. § 103(a) as unpatentable over U.S. 6,458,740 (Kasuga et al).

VII. ARGUMENT

Preface to Grounds (A) and (B)

As recited in Claim 1, the present invention is drawn to a method of producing a catalyst having a particular formula $P_aMo_bV_cCu_dX_eY_fZ_gO_h$ (wherein X, Y, Z, and a-h are as

defined), useful for subjecting methacrolein to vapor phase catalytic oxidization with molecular oxygen, the method comprising mixing

100 parts by mass of a solution or a slurry (liquid A), containing molybdenum atoms, phosphorous atoms and vanadium atoms, in which the content of ammonium species is 0 to 1.5 mol relative to 12 mol of the molybdenum atoms,

5 to 300 parts by mass of a solution or a slurry (liquid B), containing 6 to 17 mol of ammonium species relative to 12 mol of the molybdenum atoms contained in the solution A, and

a solution or a slurry (liquid C), containing an element Z,

wherein the liquid B is **mixed** with the liquid A, the liquid C or a mixture of the liquid A and the liquid C **over a period of 0.1 to 15 minutes**.

(Emphasis added.)

It is understood that the above-emphasized term means that liquid B is **added** over a period of 0.1 to 15 minutes. Once all of liquid B has been added, further stirring may occur, as can be confirmed from Example 1, for example, wherein said addition is carried out in 3 minutes, and stirring the resultant liquid was carried out for 90 minutes. In the discussion below, the terms "mixing" and "adding" are used interchangeably but are understood to be synonymous when referring to time in which mixing or adding is carried out.

Applicants describe in the specification under "Background Art," beginning at page 1, line 11, that Naito et al discloses a method for producing a catalyst for the production of methacrylic acid, wherein the catalyst produced by the method "has an insufficient yield of methacrylic acid in some cases." Applicants further describe in the specification at page 9, beginning at line 16, that in the present invention, the amount of the liquid B is 5 to 300 parts

by mass, preferably 10 to 200 parts by mass relative to 100 parts by mass of the liquid A, and that they have found that the time for mixing the liquid B with other liquids has a great influence on the performance of the catalyst to be produced and that a catalyst having a particularly high yield of methacrylic acid can be obtained by specifying the mixing time. Applicants continue that the mixing time "was not at all studied" in Naito et al, while in the present invention, the time for mixing liquid B with other liquids is 0.1 to 15 minutes.

The significance of the presently-recited mixing time for liquid B is demonstrated by the comparative data in the specification herein. The data is summarized in Table 1 at page 22 of the specification, a copy of which is in the Evidence Appendix to this Appeal Brief. As shown therein, Comparative Examples 1 and 2 are directly comparable to any of Examples 1-3 and 5, the only difference being the liquid B mixing time. As the data show, the selectivity and yield of methacrylic acid was superior using the present invention.

The above-discussed results could not have been predicted by the applied prior art.

Ground (A)

Claims 1-15 stand rejected under 35 U.S.C. § 103(a) as unpatentable over Naito et al. That rejection is untenable and should not be sustained.

Naito et al discloses liquids A, B and C, which correspond to those of the present invention. As discussed in the Preface above, Naito et al's invention is drawn to the mixing order of liquids A, B and C. A and B are first mixed together, either A into B, or B into A, or simultaneously, to form (A+B) [0019], after which (A+B) is mixed with C; i.e., (A+B)+C [0021]. In Comparative Examples 2 and 3 of Naito et al, the mixing order is changed to (A+C)+B or A+(B+C) and as a result, methacrolein conversion and methacrylic acid yield

are reduced compared to Example 1, in which the order is $(A+B)+C$. In Examples 1-4 of the present invention, the mixing order is $(A+C)+B$, similar to Comparative Example 2 of Naito et al. However, the catalysts obtained in these examples are superior in yield of methacrylic acid, in spite of the disclosure of Naito et al.

The Examiner in the Final Rejection finds that Naito et al does not limit the mixing method. The reason why the addition method is not limited is because Naito et al did not recognize any difference in addition times. However, as demonstrated herein, a superior result is provided, even if a mixing order presumed by Naito et al to be unfavorable is carried out, by adjusting the addition time of B, as claimed herein. In other words, the mixing method influences properties of the catalyst.

Since Naito et al is silent regarding mixing times, Naito et al does not present a *prima facie* case of obviousness.

Example 11 and Comparative Example 4 in the specification herein provide catalysts having the same composition as the catalyst of Example 8 of Naito et al by using the same raw materials and the same mixing order, i.e., $(A+B)+C$. Comparative Example 4 in the specification herein was conducted by the same procedure as Example 8 of Naito et al and shows the same results as Example 8 in Naito et al. In other words, Comparative Example 4 corresponds to Example 8 of Naito et al. Although Example 8 in Naito et al does not disclose the adding time, it is understood that the adding time is that disclosed for Comparative Example 4 herein. In the comparison between the results of Example 11 and Comparative Example 4, the yield of methacrylic acid increases by 1.7%. In a product like methacrylic acid produced in large quantities, even 1.7% is a very remarkable effect. Of record is a

partial translation of Example 8, and Table 1 showing conversion, selectivity and yield data for the Examples and Comparative Examples, of Naito et al.

The Examiner has not responded to the comparative data in the specification herein, discussed in the Preface and above.

Nor has the Examiner responded to Applicants' argument that Naito et al does not recognize that liquid B mixing time is a result-effective variable and therefore, the claims are patentable over Naito et al under the rationale of *In re Antonie*, 559 F.2d 618, 195 USPQ 6, 8-9 (CCPA 1977) (copy of record) (exceptions to rule that optimization of a result-effective variable is obvious, such as where the results of optimizing the variable are unexpectedly good or where the variable was not recognized to be result effective). Applicants are entitled to prevail under either of the above exceptions.

In the Final Rejection, the Examiner finds that "the mixing time is considered within the purview of ordinary skill in the art," relying on Naito et al's disclosure [0022] that the method of mixing AB with C "**is not limited**" (emphasis by the Examiner).

In reply, such disclosure actually favors Applicants, because it suggests that parameters, such as mixing time, are not previously-known result-effective variables.

In the Final Rejection, while acknowledging that Naito et al does not disclose mixing time, the Examiner finds that the lower end point of the present claims, i.e., at least 0.1 minute, i.e., 6 seconds, is within the terms of the prior art.

In reply, since Naito et al does not disclose any mixing time, mixing in the prior art could be less than the presently-recited minimum of six seconds or greater than the presently-recited maximum of 15 minutes. There is simply no basis to support the Examiner's finding.

In the Advisory Action, the Examiner states that it “cannot be ‘understood’ that the adding time disclosed for Applicants’ Comparative Example 4 corresponds to Example 8 of Naito et al; it can, at best, be only assumed that the mixing times are the same. However, the remaining elements in Applicants’ Comparative Example 4 do not correspond to elements A and AB in Example 8 of Naito et al, so the fact that the examples exhibit identical values for methacrolein conversion and methacrylic acid selectivity and yield do not necessarily mean that said identical values are a result of only the mixing time; other unknown factors could be the reason for said values.”

In reply, by comparing the description in Naito et al of Example 8 [0038], with the description in the specification herein of Example 11 and Comparative Example 4, it is clear that the same materials and amounts thereof were used. Since the conversions of methacrolein, selectivity of methacrylic acid and yield of methacrylic acid are identical for both Comparative Example 4 herein and Example 8 of Naito et al, it is indeed **understood** that the mixing time in Comparative Example 4, i.e., 30 minutes, was the mixing time for Example 8 in Naito et al.

Claims 2, 7 and 8

Claims 2, 7 and 8 are separately patentable since Naito et al neither discloses nor suggests the presently-recited amount range for their liquid C, or the presently-recited time period over which it is mixed.

Claim 5

Claim 5 is separately patentable since the catalyst made by the method of Claim 1 herein has been shown to be different from that made by the method disclosed by Naito et al.

Claim 6

Claim 6 is separately patentable since the catalyst of Claim 5 has been shown to be different from and superior to the catalyst of Naito et al., and therefore, the method of using the catalyst is different from and superior to the method of Naito et al.

Claim 10

Claim 10 is separately patentable for the reasons that Claim 2 is separately patentable combined with the reasons that Claim 5 is separately patentable.

Claim 13

Claim 13 is separately patentable for the reasons that Claim 10 is separately patentable combined with the reasons that Claim 6 is separately patentable.

For all the above reasons, it is respectfully requested that this rejection be REVERSED.

Ground (B)

Claims 1-15 stand rejected under 35 U.S.C. § 103(a) as unpatentable over U.S. 6,458,740 (Kasuga et al.). That rejection is untenable and should not be sustained.

Kasuga et al is drawn to a method for preparing a heteropolyacid catalyst containing a heteropolyacid composed of molybdophosphoric acid and/or molybdovanadophosphoric acid, or a salt of the acid, in the presence of a nitrogen-containing heterocyclic compound, which method comprises preparing an aqueous solution or dispersion which contains the nitrogen-containing heterocyclic compound, nitrate anions and ammonium ions, the ammonium content not exceeding 1.7 mols per mol of the nitrate anion content, and the ammonium ion content not exceeding 10 mols per 12 mols of the molybdenum atom content, by mixing raw materials containing the catalyst-constituting elements with a nitrogen-containing heterocyclic compound in the presence of water, then drying and calcining the same (column 2, lines 4-21).

Kasuga et al neither discloses nor otherwise suggests the presently-claimed invention. Kasuga et al contains no requirement that their catalyst-constituting raw materials be mixed in the form of three liquids, each having particular requirements, let alone that one particular liquid be mixed over a particular period of time. Nor, with regard to the catalyst *per se*, would one skilled in the art, without the present disclosure as a guide, select both (1) copper and (2) potassium, rubidium, or cesium, as the element X in Kasuga et al, and adjust “d” therein to meet the terms of presently-recited “d” and “g” herein. In sum, Kasuga et al is much further away from the presently-claimed invention than Naito et al, which Applicants assert is the closest prior art, and over which Applicants have demonstrated patentability, as discussed above.

The Examiner finds in the Final Rejection that “Example 1 of Kasuga et al disclose an exemplary embodiment wherein a solution of molybdenum and vanadium components is admixed with phosphoric acid (considered to read upon ‘liquid A’), which is then admixed

with cesium and nitrate components (considered to read upon 'liquid C'), which is then admixed with an aqueous mixture (considered to read upon 'liquid B')." This finding is clearly erroneous.

The aqueous mixture of which the Examiner finds is "liquid B" is a mixture of liquid A and liquid C (considered to read upon "mixture AC"). There is no disclosure or suggestion that the mixture AC is admixed with liquid B. Nor does Kasuga et al disclose the period of time in which liquid A is admixed with liquid C. Naturally, there is no disclosure of the mixing time of liquid B.

In addition, the ammonium ion content per 12 mols of molybdenum atoms disclosed in Kasuga et al is a final content. Although the ammonium ion content is more than 0 but not more than 10, the lowest exemplified limit of the content in Kasuga et al is 2.25 mol per 12 mol of molybdenum atoms, as shown in Example 3. On the other hand, the ammonium ion content in liquid A of the present invention is 0-1.5 mol per 12 mol of molybdenum atoms. Particularly, the content in Example 1 herein is 0. That is, in the present invention, molybdenum atom and vanadium atom are admixed with phosphoric acid in a lower ammonium ion content, preferably in the absence of ammonium ion.

While the Examiner finds that Applicants' argument that Kasuga et al is much further away from the presently-claimed invention than Naito et al, "does not detract said reference [Kasuga et al] from reading upon the instantly claimed invention," the Examiner has not responded to Applicants' argument that Naito et al is the closest prior art, and over which Applicants have demonstrated patentability, as discussed above. In addition, if Kasuga et al reads on the present invention, why did the Examiner not reject under 35 U.S.C. § 102(b)?

In both the Final Rejection and Advisory Action, the Examiner finds that the presently-recited minimum mixing time of six seconds is within the terms of Kasuga et al.

Applicants' reply is the same as the reply discussed above with regard to Naito et al.

Claim 2

Claim 2 is separately patentable since Kasuga et al neither discloses nor suggests the presently-recited amount range for their liquid C, or the presently-recited time period over which it is mixed.

Claim 3

Claim 3 is separately patentable since Kasuga et al neither discloses nor suggests employing a solution or a slurry which contains substantially no phosphorous, molybdenum, vanadium, copper, element X, element Y or element Z to make their catalyst.

Claim 4

Claim 4 is separately patentable since Kasuga et al neither discloses nor suggests employing a solution or a slurry which contains substantially no phosphorous, molybdenum, vanadium, copper, element X, element Y or ammonium species to make their catalyst.

Claim 5

Claim 5 is separately patentable since Kasuga et al neither discloses nor suggests the claimed catalyst.

Claim 6

Claim 6 is separately patentable since Kasuga et al neither discloses nor suggests a method of producing methacrylic acid using the recited catalyst.

Claim 7

Claim 7 is separately patentable for the reasons that Claim 2 is separately patentable combined with the reasons that Claim 3 is separately patentable.

Claim 8

Claim 8 is separately patentable for the reasons that Claim 2 is separately patentable combined with the reasons that Claim 4 is separately patentable.

Claim 9

Claim 9 is separately patentable for the reasons that Claim 3 is separately patentable combined with the reasons that Claim 4 is separately patentable.

Claim 10

Claim 10 is separately patentable for the reasons that Claim 2 is separately patentable combined with the reasons that Claim 5 is separately patentable.

Claim 11

Claim 11 is separately patentable for the reasons that Claim 3 is separately patentable combined with the reasons that Claim 5 is separately patentable.

Claim 12

Claim 12 is separately patentable for the reasons that Claim 4 is separately patentable combined with the reasons that Claim 5 is separately patentable.

Claim 13

Claim 13 is separately patentable for the reasons that Claim 10 is separately patentable combined with the reasons that Claim 6 is separately patentable.

Claim 14

Claim 14 is separately patentable for the reasons that Claim 11 is separately patentable combined with the reasons that Claim 6 is separately patentable.

Claim 15

Claim 15 is separately patentable for the reasons that Claim 12 is separately patentable combined with the reasons that Claim 6 is separately patentable.

For all the above reasons, it is respectfully requested that this rejection be
REVERSED.

VIII. CONCLUSION

For the above reasons, it is respectfully requested that all the rejections still pending in the Final Rejection be REVERSED.

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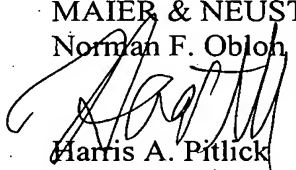
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CLAIMS APPENDIX

Claim 1: A method of producing a catalyst having a composition of the following formula (1), wherein said catalyst is useful for production of methacrylic acid by subjecting methacrolein to vapor phase catalytic oxidization with molecular oxygen,

said method comprising mixing

100 parts by mass of a solution or a slurry (liquid A), containing molybdenum atoms, phosphorous atoms and vanadium atoms, in which the content of ammonium species is 0 to 1.5 mol relative to 12 mol of the molybdenum atoms,

5 to 300 parts by mass of a solution or a slurry (liquid B), containing 6 to 17 mol of ammonium species relative to 12 mol of the molybdenum atoms contained in the solution A, and

a solution or a slurry (liquid C), containing an element Z,

wherein the liquid B is mixed with the liquid A, the liquid C or a mixture of the liquid A and the liquid C over a period of 0.1 to 15 minutes:



wherein P, Mo, V, Cu and O represent phosphorous, molybdenum, vanadium, copper and oxygen, respectively; X represents at least one element selected from the group consisting of antimony, bismuth, arsenic, germanium, zirconium, tellurium, silver, selenium, silicon, tungsten and boron; Y represents at least one element selected from the group consisting of iron, zinc, chromium, magnesium, tantalum, cobalt, manganese, barium, gallium, cerium and lanthanum; Z represents at least one element selected from the group consisting of potassium, rubidium and cesium; a, b, c, d, e, f, g and h represent an atomic ratio of each element, and when b=12, a=0.5 to 3, c=0.01 to 3, d=0.01 to 2, e=0 to 3, f=0 to 3 and g=0.01 to 3, and h

represents an atomic ratio of oxygen necessary for satisfying the valence of each of the above-mentioned components.

Claim 2: The method according to claim 1, wherein 5 to 100 parts by mass of the liquid C is mixed with the liquid A, or a mixture of the liquid A and the liquid B over 0.1 to 30 minutes.

Claim 3: The method according to claim 1, wherein the liquid B is a solution or a slurry which contains substantially no phosphorous, molybdenum, vanadium, copper, element X, element Y or element Z.

Claim 4: The method according to claim 1, wherein the liquid C is a solution or a slurry which contains substantially no phosphorous, molybdenum, vanadium, copper, element X, element Y or ammonium species.

Claim 5: A catalyst for use in the production of methacrylic acid, which is produced by the method claimed in claim 1.

Claim 6: A method of producing methacrylic acid, comprising, subjecting methacrolein to vapor phase catalytic oxidization with molecular oxygen in the presence of the catalyst for producing methacrylic acid as claimed in claim 5.

Claim 7: The method according to claim 2, wherein the liquid B is a solution or a slurry which contains substantially no phosphorous, molybdenum, vanadium, copper, element X, element Y or element Z.

Claim 8: The method according to claim 2, wherein the liquid C is a solution or a slurry which contains substantially no phosphorous, molybdenum, vanadium, copper, element X, element Y or ammonium species.

Claim 9: The method according to claim 3, wherein the liquid C is a solution or a slurry which contains substantially no phosphorous, molybdenum, vanadium, copper, element X, element Y or ammonium species.

Claim 10: A catalyst for use in the production of methacrylic acid, which is produced by the method claimed in claim 2.

Claim 11: A catalyst for use in the production of methacrylic acid, which is produced by the method claimed in claim 3.

Claim 12: A catalyst for use in the production of methacrylic acid, which is produced by the method claimed in claim 4.

Claim 13: A method of producing methacrylic acid, comprising, subjecting methacrolein to vapor phase catalytic oxidization with molecular oxygen in the presence of the catalyst for producing methacrylic acid as claimed in claim 10.

Claim 14: A method of producing methacrylic acid, comprising, subjecting methacrolein to vapor phase catalytic oxidization with molecular oxygen in the presence of the catalyst for producing methacrylic acid as claimed in claim 11.

Claim 15: A method of producing methacrylic acid, comprising, subjecting methacrolein to vapor phase catalytic oxidization with molecular oxygen in the presence of the catalyst for producing methacrylic acid as claimed in claim 12.

EVIDENCE APPENDIX

Table 1 at page 22 of the specification.

Partial translation of Example 8, and Table 1 showing conversion, selectivity and yield data for the Examples and Comparative Examples, of Naito et al, filed January 4, 2007.

Table 1

	Amount of ammonium species in liquid B ¹⁾	Mass ²⁾ (part)		Mixing time (min)		Conversion of methacrolein (%)	Selectivity of methacrylic acid (%)	Yield of methacrylic acid (%)
		Liquid B	Liquid C ³⁾	Liquid B	Liquid C ³⁾			
Example 1	10.0	12.64	13.45	3.0	5.5	82.5	87.6	72.3
Example 2	10.0	12.64	13.45	8.0	5.5	83.0	87.0	72.2
Example 3	10.0	12.64	13.45	13.0	5.5	82.7	87.5	72.4
Example 4	10.0	12.64	13.45	13.0	25.0	82.0	87.9	72.1
Example 5	10.0	12.64	13.45	1.0	5.5	81.3	88.0	71.5
Example 6	7.5	9.48	13.45	1.5	5.5	81.1	87.9	71.3
Example 7	14.0	17.69	13.45	4.0	5.5	82.0	87.5	71.8
Example 8	10.0	12.64	13.45	3.0	0.8	80.8	88.1	71.2
Comparative Example 1	10.0	12.64	13.45	0.05	5.5	81.0	85.7	69.4
Comparative Example 2	10.0	12.64	13.45	20.0	5.5	81.8	84.0	68.7
Example 9	11.5	10.96	8.30 5.54	2.0	2.5 1.5	84.5	86.1	72.8
Example 10	11.5	10.96	8.30 5.54	2.0	13.0 0.6	85.0	84.5	71.8
Comparative Example 3	11.5	10.96	8.30 5.54	40.0	2.5 1.5	82.5	84.3	69.5
Example 11	9.2	78.04	15.53	2.5	7.5	81.1	85.8	69.6
Comparative Example 4	9.2	78.04	15.53	30.0	35.0	81.2	83.6	67.9

1) Number of moles relative to 12 mol of molybdenum in liquid A

2) Mass based on 100 parts by mass of liquid A

3) Of the two lines, upper line indicating value of liquid Cl, lower line indicating value of liquid CII

[0038] [Example 8]

To 400 parts of pure water were added 100 parts of molybdenum trioxide, 8.88 parts of 85% by mass phosphoric acid, 4.74 parts of ammonium metavanadate and 4.11 parts of 60% aqueous arsenic acid solution and stirring was conducted in an autoclave under saturated vapor at 120°C for 3 hours to prepare liquid A. The amount of ammonium species contained in the liquid A was 0.7 mol relative to 12 mols of the molybdenum atoms contained in the liquid A. After cooling the liquid A to 60°C, a solution in which 37.0 parts of ammonium carbonate was dissolved in 80 parts of pure water, which is liquid B was added dropwise to the liquid A and then stirred for 15 minutes to obtain AB mixed solution. The amount of ammonium species in the AB mixed solution was 14 mols relative to 12 mols of the molybdenum atoms. Then, a solution in which 1.40 parts of cupric nitrate was dissolved in 10 parts of pure water, a solution in which 2.34 parts of ferric nitrate was dissolved in 10 parts of pure water and 1.00 part of cerium oxide were added in sequence and with stirring the mixture, liquid C in which 18.0 parts of cesium bicarbonate was dissolved in 30 parts of pure water was added dropwise and stirring was conducted for 15 minutes to prepare a slurry. The slurry thus obtained was heated to 101°C and subjected to evaporation to dryness with stirring. The obtained solid was dried at 130°C for 16 hours. The dried product was subjected to pressing, calcined under nitrogen stream at 400°C for 5 hours and further calcined under air stream at 340°C for 10 hours to obtain a catalyst whose composition was $P_{1.3}Mo_{12}V_{0.7}Cu_{0.1}Fe_{0.1}As_{0.3}Ce_{0.1}Cs_{1.6}$. The results of the reaction conducted using this catalyst in the same manner as in Example 1, except for changing the reaction temperature to 290°C, are shown in Table 1.

[0040]

[Table 1]

	Adding order ¹⁾	Ratio of Ammonium species		Conversion of MAL ³⁾ (%)	Selectivity of MAA ⁴⁾ (%)	Single Current Yield of MAA ⁴⁾ (%)
		Liquid A	AB mixed solution ²⁾			
Ex. 1	(A+B)+C		11.0	83.6	82.4	68.9
Ex. 2	(A+B)+C		6.0	83.0	82.8	68.7
Ex. 3	(A+B)+C		15.0	83.3	82.6	68.8
Ex. 4	(A+B)+C		4.0	80.5	83.1	66.9
Ex. 5	(A+B)+C		20.0	79.8	83.4	66.6
Ex. 6	(A+B)+C		11.0	81.5	82.5	67.2
Comp. Ex.1	(A+B)+C	4.0	7.0	78.6	83.0	65.2
Comp. Ex.2	(A+C)+B		11.0	77.7	83.2	64.6
Comp. Ex.3	A+(B+C)		11.0	78.3	83.0	65.0
Ex. 7	(A+B)+C	0.7	13.3	80.3	83.5	67.1
Ex. 8	(A+B)+C	0.7	13.3	81.2	83.6	67.9
Comp. Ex.4	(A+B)+C	0.7	13.3	76.5	83.4	63.8

1) Means of code in the column of adding order are as follows:

(A+B)+C: Liquid B is added to liquid A and then liquid C is added thereto.

(A+C)+B: Liquid C is added to liquid A and then liquid B is added thereto.

A+(B+C): A mixed solution of liquid B and liquid C is added to liquid A.

(A+B+C): Liquids A, B and C are mixed at one time.

- 2) In the case other than that the adding order is (A+B)+C, ratio of ammonium species of AB mixed solution is that of the mixed solution including liquid C.
- 3) MAL represents methacrolein.
- 4) MAA represents methacrylic acid.

RELATED PROCEEDINGS APPENDIX

None.